PATENT COOPERATION TREATY

PCT

INTERNATIONAL PRELIMINARY EXAMINATION REPORT
(PCT Article 36 and Rule 70)

Applicant's or agent's file reference	FOR FURTHER ACTION	See Notification of Transmittal of International Preliminary Examination Report (Form PCT/IPEA/416)
SMX 2014.1		
International application No. PCT/US 03/06613	International filing date *(day/month/year)* 06.03.2003	Priority date *(day/month/year)* 06.03.2002
International Patent Classification (IPC) or both national classification and IPC H01M4/92		
Applicant SYMYX TECHNOLOGIES, INC. et al.		

1. This international preliminary examination report has been prepared by this International Preliminary Examining Authority and is transmitted to the applicant according to Article 36.

2. This REPORT consists of a total of 8 sheets, including this cover sheet.

   ☒ This report is also accompanied by ANNEXES, i.e. sheets of the description, claims and/or drawings which have been amended and are the basis for this report and/or sheets containing rectifications made before this Authority (see Rule 70.16 and Section 607 of the Administrative Instructions under the PCT).

   These annexes consist of a total of 5 sheets.

3. This report contains indications relating to the following items:

   - I ☒ Basis of the opinion
   - II ☐ Priority
   - III ☐ Non-establishment of opinion with regard to novelty, inventive step and industrial applicability
   - IV ☐ Lack of unity of invention
   - V ☒ Reasoned statement under Rule 66.2(a)(ii) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement
   - VI ☐ Certain documents cited
   - VII ☐ Certain defects in the international application
   - VIII ☐ Certain observations on the international application

Date of submission of the demand	Date of completion of this report
03.10.2003	16.04.2004
Name and mailing address of the international preliminary examining authority: European Patent Office D-80298 Munich Tel. +49 89 2399 - 0 Tx: 523656 epmu d Fax: +49 89 2399 - 4465	Authorized Officer Hintermaier, F Telephone No. +49 89 2399-7063

INTERNATIONAL PRELIMINARY EXAMINATION REPORT

International application No. PCT/US 03/06613

I. Basis of the report

1. With regard to the **elements** of the international application *(Replacement sheets which have been furnished to the receiving Office in response to an invitation under Article 14 are referred to in this report as "originally filed" and are not annexed to this report since they do not contain amendments (Rules 70.16 and 70.17))*:

Description, Pages

1-37 as originally filed

Claims, Numbers

1-35 received on 14.10.2003 with letter of 10.09.2003

Drawings, Sheets

1/2-2/2 as originally filed

2. With regard to the **language**, all the elements marked above were available or furnished to this Authority in the language in which the international application was filed, unless otherwise indicated under this item.

   These elements were available or furnished to this Authority in the following language: , which is:

   ☐ the language of a translation furnished for the purposes of the international search (under Rule 23.1(b)).

   ☐ the language of publication of the international application (under Rule 48.3(b)).

   ☐ the language of a translation furnished for the purposes of international preliminary examination (under Rule 55.2 and/or 55.3).

3. With regard to any **nucleotide and/or amino acid sequence** disclosed in the international application, the international preliminary examination was carried out on the basis of the sequence listing:

   ☐ contained in the international application in written form.

   ☐ filed together with the international application in computer readable form.

   ☐ furnished subsequently to this Authority in written form.

   ☐ furnished subsequently to this Authority in computer readable form.

   ☐ The statement that the subsequently furnished written sequence listing does not go beyond the disclosure in the international application as filed has been furnished.

   ☐ The statement that the information recorded in computer readable form is identical to the written sequence listing has been furnished.

4. The amendments have resulted in the cancellation of:

   ☐ the description, pages:

   ☐ the claims, Nos.:

   ☐ the drawings, sheets:

5. ☐ This report has been established as if (some of) the amendments had not been made, since they have been considered to go beyond the disclosure as filed (Rule 70.2(c)).

   (Any replacement sheet containing such amendments must be referred to under item 1 and annexed to this report.)

6. Additional observations, if necessary:

V. Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement

1. Statement

Novelty (N)	Yes:	Claims	7 - 10, 15 - 35
	No:	Claims	1 - 6, 11 - 14
Inventive step (IS)	Yes:	Claims	
	No:	Claims	7 - 10, 15 - 35
Industrial applicability (IA)	Yes:	Claims	1 - 35
	No:	Claims	

2. Citations and explanations

   see separate sheet

Re Item V

Reasoned statement under Rule 66.2(a)(ii) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement

1. Reference is made to the following documents:

D1: US-A-4 100 180 (ICHIKAWA YATARO ET AL) 11 July 1978 (1978-07-11)
D2: PATENT ABSTRACTS OF JAPAN vol. 001, no. 131 (C-029), 28 October 1977 (1977-10-28) & JP 52 084193 A (TEIJIN LTD), 13 July 1977 (1977-07- 13)
D3: US-A-4 536 259 (ENDOH EIJI ET AL) 20 August 1985 (1985-08-20)
D4: US-A-4 298 462 (ANTOS GEORGE J) 3 November 1981 (1981-11-03)
D5: DE 17 96 043 A (SIEMENS AG) 2 March 1972 (1972-03-02)
D6: EP-A-0 129 399 (ENGELHARD CORP) 27 December 1984 (1984-12-27)
D7: EP-A-0 899 348 (EXXON RESEARCH ENGINEERING CO) 3 March 1999 (1999-03-03)
D8: PATENT ABSTRACTS OF JAPAN vol. 1997, no. 12, 25 December 1997 (1997-12-25) & JP 09 206597 A (AQUEOUS RES:KK), 12 August 1997 (1997-08-12)

2. Prior art and novelty (Article 33(2) PCT).

2.1. D1 teaches a solid catalyst for the selective hydrogenation of aldehyde groups (abstract). In Example 6 platinum oxide is treated in an autoclave with iron sulfate and zinc acetate in the presence of hydrogen. The resulting catalyst has an iron retention of 43.6 % per weight and a zinc retention of 5.0 % per weight. Since the preparation of the catalyst was carried out in a hydrogen atmosphere it is concluded that the catalyst consists essentially only of the metals Pt, Zn and Fe. Therefore, the Pt content must be 51.4 wt. %. Since in an alloy not all metallic constituents must have an oxidation state of 0, the catalyst is considered to constitute an alloy. Considering the atomic masses of these metals this corresponds to the following atomic percentages: Pt: 23 %, Zn: 6.9 % and Fe: 70 %. Thus, claims 1 - 6, 11, and 12 lack novelty over D1.

2.2. Also D2 discloses a solid catalyst for the selective hydrogenation of aldehyde groups, the catalyst comprising Pt oxide carrying Fe and Zn. Since the catalyst is used in an H_2 atmosphere at least a part of the metals will be reduced to the metal state,

thereby forming an alloy. The composition in the surface layer is 1.5 - 70 wt. % Pt and 0.5 - 8 wt. % Zn. Considering the atomic masses for the metals, D2 is prejudicial for novelty of claims 1 - 6 and 11 - 14 of the present application.

2.3. D3 discloses a cathode having a high durability and low hydrogen overvoltage, for example for an application in electrolysis (col. 1, line 6 - 16). The electrode consists of an electrically conductive substrate onto which an alloy is coated (col. 6 line 20, - col. 8, line 44). The alloy consist of a noble metal, a metal which may be removed fully or partially by dissolution and Ni or Co or a mixture of both (col. 3, line 43 - 68). Comparative example 6 (Table 2) comprises a Pt-Zn-Ni alloy having 10 wt. % Pt, 10 wt. % Zn and 80 wt. % Ni. This alloy is considered as a catalyst, since it contains Pt. However, the composition of comparative example 6 lays out of the scope of claim 1, since its Pt content is too low. Therefore, D3 is not prejudicial for novelty.

2.4. D4 describes a catalyst for the conversion of hydrocarbons, which comprises in combination a Pt group component, a Ni component, a Zn component, a halide component and a porous carrier material (abstract). The catalyst is applied to the carrier by impregnation. In Example II (col. 21) a catalyst is described having an atomic ratio of Ni:Pt = 8.9:1 and of Zn:Pt = 4:1. This corresponds to a composition of 7 at. % Pt, 29 at. % Zn and 64 at. % Ni. Preparation of the catalyst ends with a reduction step during which metallic Pt and Ni are formed (col. 14, line 5 - 39, and col. 19, line 51, - col. 20, line 49), which means that the catalyst is an alloy. D4 is not novelty-destroying for any of the claims, because the disclosed alloys do not have a Pt concentration between 10 - 80 at. %.

2.5. D5 discloses a catalyst electrode for a fuel cell, which has a high activity and a good adhesion on its electrically conducting carrier (page 2, line 18 - 20). As the fuel hydrazine is mentioned. Onto the carrier, which may consist of carbon, graphite or a metal, first a layer of Raney-Ni is deposited, for example by electroplating from a $NiSO_4/ZnSO_4/Na(CH_3COO)$ solution (page 2, line 21, - page 4, line 1). Following, a noble metal is deposited (page 4, line 16 - 21), with the elements Pt, Ir, Ru, Rh, Pd and Os explicitly being mentioned. In the process of activation a part of the inactive component of the Raney alloy, i.e. a part but not all of the Zn, is removed by dissolution in KOH so that there is a sufficient amount of Zn remaining for the following electroless deposition of the noble metal. D4 is not novelty-destroying for any of the claims, because no Pt contents are mentioned in numbers.

2.6. D6 teaches a supported Pt-Fe alloy electrocatalyst, which consists of 17 - 42 at. % Fe with the balance being Pt. The catalyst is preferably supported on carbon powder or the like and shows a greater activity for oxygen reduction and better resistance to sintering (abstract). The metals are applied by impregnating the carrier. D6 is not detrimental for novelty, because it does not disclose a combination of Pt, Fe and Zn.

2.7. D7 describes a CO-tolerant Pt-Zn alloy for use in a fuel cell electrode. In D7 particles of carbon-supported Pt (Vulcan XC-72®) are impregnated with a $Zn(NO_3)_2$ solution and subsequently reduced in H_2 (col. 5, line 1 - 20). D7 is not detrimental for novelty, because it does not disclose a combination of Pt, Zn and one or both of Fe and Ni.

2.8. D8 discloses an alloy of Pt and Fe to be used as a catalyst in a fuel cell. It is stated that the catalyst is highly active. However, Zn is not included in the catalyst composition.

2.9. Independent claims 16, 19, 22, 31 and 35 are considered to be novel.

3. Inventive step (Article 33(3) PCT).

3.1. Claims 7 - 10 and 15 are not inventive, because they differ from the non-novel subject-matter of claims 5, 6 and 11 - 14 only with respect to the numerical ranges of the composition of the catalyst and there is no further non-obvious technical effect taught in the present application which would be correlated with the ranges of these claims.

3.2. As to the remaining claims 16 - 34 only the question needs to be discussed whether the use of an alloy composition, which combines Pt, Zn and one or both of Fe and Ni as an electrocatalyst in an electrochemical reactor, such as a fuel cell, is inventive, since there are no further technical features seen in these claims which are not known from the prior art:

3.2.1. In the prior art (e.g. D6, D7) it is known to have Pt or other noble metals supported by conductive particles, e.g. made from carbon or graphite (claims 16 - 21).

3.2.2. Claims 24 - 26 try to define a PEM fuel cell by the fuel. However, a regular PEM fuel cell is considered to work with H_2, a hydrocarbon-based fuel and MeOH, e.g. the

fuel cells of D6 and D7.
3.2.3. With respect to claims 27 - 30 there are no special technical effects taught in the present application related to the location of the catalyst. For example in D7 the catalyst is applied to a carbon cloth, which forms part of the electrode.
3.2.4. A method for the electrochemical conversion of a hydrogen-containing fuel (claim 31) is already known from the art (e.g. D5 - D8). The skilled person would also consider H_2 or hydrocarbons (claims 32 - 33) as fuels in these methods.

3.3. Hence, the question remains whether the use of an alloy which combines Pt, Zn and one or both of Fe and Ni in a fuel cell is inventive. A criteria for inventiveness is the occurrence of a **non-obvious technical effect**. Non-obvious means that this effect is not derivable from the prior art, e.g. by extrapolation of effects taught in a prior art document or by due combination of the teachings of two or more prior art documents. In the present application the different catalyst compositions were only examined on their activity. However, the results presented are not conclusive and even in part contradictory (see point 2 above, especially 2.4.). Hence, **there is no new surprising technical effect taught**, which an electrocatalyst combining Pt, Zn and one or both of Fe and Ni would have over state-of-the-art electrocatalysts. Therefore, claims 16 - 34 are not considered to be inventive.
This comment is also valid, if novelty over D1 and D2 would be established by specifying that all metals of claim 1 must be in an oxidation state of 0.
Finally it is to be noted that the present application does not specify, which technical problem it seeks to overcome.

3.4. Moreover, the skilled person who faces the problem of improving resistance to poisoning and who wants to improve the activity of the electrocatalyst of a fuel cell (subjective problem of the Applicant, see 2.1., above) could and would simply combine documents D7 and D8, which address already the problem of an improvement of the CO tolerance and of the activity, respectively.

3.5. Claim 35 is not considered inventive, since there are no non-obvious technical effects taught, which an unsupported catalyst would have over the supported catalysts of D1 or D2.

4. Industrial applicability (Article 33(4) PCT).

Claims 1 - 35 fulfill the requirement of industrial applicability, since subject-matter of present application can be made or used (in a technological sense) in industry (Article 33(4) PCT).

5. Clarity

5.1. The term "about" in claims 1, 3 - 15, which is used for the specification of the composition of the alloy, is vague and leaves the reader in doubt about which range is intended to be claimed.

5.2. The ranges of claim 12 don't fit together: in case the catalyst has an upper Fe portion of 80 at. %, the sum of the lower portions of Pt and Zn can only be 20 at. % and not 25 at. %, since the mentioned constituents cannot add up to more than 100%. Page 8, line 30 -32, however states a catalyst composition which has he same ranges for Pt and Zn as in claim 12 and which has a range for iron of 10 - 70 at. %.

5.3. Units like "Angstroms" (page 12, line 1 - 11, page 15, line 12 - 13, page 18, line 28, - page 19, line 4, page 33, line 28 - 30) do not meet the requirements of Rule 10 PCT, which requests use of SI units.

6. Documents D5 - D7 should be cited as prior art to comply with the provisions of Rule 5.1.a.ii PCT.